



PATENT--FEE

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ASHOKE SENGUPTA ET AL.) paper is being deposited
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Examiner: William K. Cheung) Commissioner for Patents
) P.O. Box 1450
) Alexandria, VA 22313-1450
)
) Dated: January 17, 2006
)
)
) James J. Napoli _____
) James J. Napoli
) Registration No. 32,361
) Attorney for Applicants

**DECLARATION OF ASHOKE K. SENGUPTA
UNDER 37 C.F.R. §1.132**

Mail Stop Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

Sir:

NOW COMES ASHOKE K. SENGUPTA, Declarant herein, and states as follows:

1. I am a co-inventor of the invention disclosed and claimed in the above-identified patent application.

2. I am presently employed by AMCOL International Corporation, Arlington Heights, Illinois, assignee of the above-identified patent application. My present title is Senior Principal Scientist. I have been employed by AMCOL since 2000, engaged in research

for developing sunscreen additives, polymer-modified clay-based thickeners for nonpolar solvents, deposition-aids for depositing oily/hydrophobic actives from cleansing compositions, and polymeric and clay-based coagulation aids for wastewater.

3. Prior to my employment at AMCOL, I was employed at Borregaard, a Norwegian specialty chemical company, as a Senior Research Scientist, involved in developing technologies for polymeric dispersants and surface modifiers used in concrete and mining operations, emulsifiers, and coagulation-aids.

4. I earned a B.S. (Honors) in chemical engineering from Jadavpur University, India (1987) and a Ph.D. in chemical engineering from Tulane University, New Orleans, LA (1994), studying colloidal phenomena related to dispersion stability, electrokinetic transport of colloids, polymeric emulsifiers, and stability of dispersions in the presence of polymers.

5. I have conducted research in numerous areas involving dispersion/surface-modification technologies. I have published in the *Journal of Colloid and Interface Science* and the *Journal of the Chemical Society, Faraday Transactions*, and have presented at numerous professional and scientific conferences. I am a named inventor in several patents and patent publications.

6. I have read and understand the Office Action dated September 19, 2005, which was issued in connection with U.S. Patent Application Serial No. 10/626,009. I also have read and understand the following publications cited by the examiner in U.S.S.N.

10/626,009: Finlayson U.S. Patent No. 4,208,218 ('218) and Wong et al. U.S. Patent No. 4,830,945 ('945).

7. Claims 1-10, 12-18, and 34-37 of U.S.S.N. 10/626,009 were rejected as being obvious over the '218 patent in view of the '945 patent because the '218 patent teaches surface modifying a layered silicate material with a quaternary ammonium compound, and because the '945 patent teaches resin particles with adsorbed amphipathic copolymer moieties having an ability to act as steric stabilizers. The examiner also states that no comparative data showing unexpected results over the cited references have been provided.

8. After reading the cited references, and based on my training and experience, with support from the comparative data set forth in paragraphs 16, 18, 20, and 23, the pending claims would not have been obvious to a person of ordinary skill over the '218 and '945 patents. Prior to discussing the cited references and the comparative data, paragraph 9 below discusses specific drawbacks of the prior art, which are overcome by the present invention, followed by an overview of the claimed invention in paragraph 10.

9. Organophilic clays (layered silicates), known in the art as thickeners for hydrophobic liquids, traditionally have been produced by surface modifying the clay surface using long chain (C_{14-22}) quaternary ammonium compounds (QAC). For health, odor, and skin irritation-related issues, these QACs, especially those derived from tallow and/or containing aromatic groups, preferably are avoided in formulating personal care and cosmetic products. Both the QACs claimed in the '218 patent contain benzyl groups (aromatic), one of which

even is derived from tallow. A critical object of the present invention was to be able to produce organoclay-based thickeners for hydrophobic liquids, without having to use QACs as the clay surface modifier.

10 Accordingly, the present claims are directed to a composition capable of thickening hydrophobic liquids, comprising (a) a layered silicate material, for example, smectite clay, having plate-like particles (referred to as platelets), wherein the surfaces of the layered silicate are modified by (b) an amphipathic copolymer. The amphipathic copolymer comprises (i) a first comonomer that when homopolymerized generates a hydrophilic polymer that is insoluble in hydrophobic liquids and (b) a second comonomer that when homopolymerized generates a hydrophobic polymer that is soluble in a hydrophobic liquid. Unexpectedly, it has been found that this amphipathic copolymer is capable of delaminating/exfoliating (i.e., separating clay platelets across their face-surfaces from stacks of tightly aggregated platelets) clay platelets in a hydrophobic liquid, in a manner such that the delaminated/exfoliated clay platelets then can thicken the hydrophobic liquid medium in which they are dispersed. As discussed in paragraphs 11 and 12, dispersions tend to exhibit thinning, rather than thickening, when the dispersed particles are surface-treated with polymeric surface modifiers. Accordingly, while thinning is the norm for polymeric surface modifiers, the present invention provides the opposite, i.e., thickening.

11. A wide use of polymers, both homopolymers and copolymers, is as a dispersing agent or dispersant. A polymeric dispersing agent enables sus-

suspended particles to repel one another, i.e., to counteract their inherent mutual attraction, by appropriately modifying the particle surface. Such interparticle repulsion, known as steric repulsion, stabilizes the particles against agglomeration or flocculation, and hence the term "steric stabilizer" for these dispersants.

12. With respect to the rheological effects of a dispersing agent, a dispersing agent generally tends to reduce the viscosity of concentrated dispersions by minimizing agglomeration of suspended particles. More particularly, suspended particles are said to be "agglomerated" when they remain so strongly attracted to their surrounding particles that they cannot move independently of these neighboring particles under an applied force, even though they may be physically separated from one another. "Moving independently" means that any given particle is able to move in any direction, regardless of the direction in which its neighboring particles move at any instant of time. In effect, the particle-to-particle attraction translates to particulate structures formed in the dispersion, within which the suspending liquid remains partially or wholly entrapped. As long as an externally applied force (e.g., a shear force) is unable to break apart these particulate structures, and thus release the entrapped suspending liquid, the dispersion remains viscous. The entrapped liquid is no longer available to impart fluidity for the dispersion, and hence the increased viscosity. A dispersant's function is to minimize particle-to-particle "bonds" or attractions that result in particulate structures. In gen-

eral, the steric stabilizers, namely the polymeric dispersants, are particularly effective in reducing interparticle attraction. Therefore, the expected result of using steric stabilizers is a reduction in dispersion viscosity.

13. The '945 patent is directed to a photographic developer. The developer contains certain resin particles having amphipathic copolymeric moieties attached thereto. As described in the '945 patent, these moieties act to prevent flocculation, i.e., serve as steric stabilizers. Accordingly, the '945 patent teaches that the amphipathic copolymers disclosed therein can act as steric stabilizers. Based on the discussion in paragraph 12, the teachings of the '945 patent would prompt persons skilled in the art to expect these copolymers to render smectite clay dispersions nonviscous rather than viscous. For this reason, the teachings of the '945 patent would not encourage using the copolymers disclosed therein as a clay surface modifier in compositions wherein smectite clays thicken hydrophobic liquids. Furthermore, given some of the examples of the copolymers disclosed therein, the '945 patent seemingly does not require the surface-modifying copolymer to comprise a hydrophilic comonomer and a hydrophobic comonomer, as required by the present claims. For example, copolymers such as poly(butyl methacrylate-co-methyl methacrylate), poly(ethylene-co-methacrylic acid), poly(ethylene-co-vinyl acetate) disclosed in the '945 patent essentially comprise of only hydrophobic comonomers.

14. To demonstrate the new and unexpected benefits provided by the compositions of the present

invention, twelve dispersion samples, summarized in Table I of paragraph 16, were prepared under my supervision. In each sample, 5 wt% (4.65 wt% on moisture-free basis, based on the whole dispersion) of a purified smectite clay was dispersed in the indicated solvent in the presence of a copolymeric surface modifier, and a polar activator, a 90.2:9.8 mixture of propylene carbonate and water, added at a dosage of about 2.55 wt%. The surface modifier dosage was 5 wt% for all samples, except for Sample No. 3 which contained 12.5 wt% of a mixture of copolymers of the present invention. Note that additional dispersion samples (not presented herein for the sake of brevity), using octyl stearate as the hydrophobic solvent, were prepared under my supervision, using three copolymers, namely, poly(isoprene-graft-maleic anhydride), poly(butyl methacrylate-co-methyl methacrylate), and poly(ethylene-co-vinyl acetate) of the '945 patent, and a claimed copolymer (PEG 30 dipolyhydroxystearate) of the present invention, wherein the clay dosage was the same as, but the copolymer dosage was half (i.e., 2.5 wt%) of that in the corresponding samples summarized in Table I of paragraph 16.

15. It is evident from Table I below that Sample Nos. 1-3, utilizing copolymers of the present invention, exhibited substantial thickening of various hydrophobic solvents. Note that, absent the copolymer, the clay alone failed to thicken the solvents, while the solutions containing a claimed copolymer alone were hardly any more viscous than the solvents alone, without the clay. In contrast, Sample Nos. 4-12 containing copolymers outside the scope of the claimed copolymers,

including mostly the copolymers of the '945 patent, did not exhibit any thickening. Within an hour after their preparation, these dispersions separated into two phases, a solvent-rich phase over a solid-rich phase of settled clay particles. Similar results, as noted above, were observed even when the copolymer dosage was reduced to 2.5 wt%.

16.

Table I

Sample No.	Surface modifier	% Clay	Solvent	Brookfield Viscosity/Comment
1	PEG 30 dipolyhydroxy-stearate (a copolymer of the present invention)	5	Octyl stearate (ester)	184,000 cps @ 1 rpm 88,000 cps @ 5 rpm
2	PEG 30 dipolyhydroxy-stearate (a copolymer of the present invention)	5	Isododecane (hydrocarbon)	120,000 cps @ 1 rpm 32,000 cps @ 5 rpm
3	Cetyl PEG/PPG-10/1 polydimethylsiloxane and PEG-30 dipolyhydroxy-stearate (a copolymer mixture of the present invention)	5	Cyclomethicone (silicone)	30,000 cps @ 1 rpm 12,000 cps @ 5 rpm
4	Polyglyceryl-2 dipolyhydroxystearate	5	Octyl stearate	No thickening
5	Poly(butyl methacrylate - co-methyl methacrylate) (a copolymer from the first set in the '945 patent)	5	Octyl stearate	No thickening
6	Poly(ethylene-co-methacrylic acid) (a copolymer from the second set in the '945 patent)	5	Octyl stearate	No thickening
7	Poly(ethylene-co-vinyl acetate) (a copolymer from the second set in the '945 patent)	5	Octyl stearate	No thickening
8	Poly(isoprene-graft-maleic anhydride) (a copolymer from the '945 graft copolymers)	5	Octyl stearate	No thickening
9	Poly(butyl methacrylate-co-methyl methacrylate)	5	Isododecane	No thickening
10	Poly(ethylene-co-methacrylic acid)	5	Isododecane	No thickening
11	Poly(ethylene-co-vinyl acetate)	5	Isododecane	No thickening
12	Poly(ethylene-graft-maleic anhydride)	5	Isododecane	No thickening

17. To demonstrate further the unexpected properties of the compositions of the present invention, ten dispersion samples were prepared under my

supervision, which are summarized in Table II of paragraph 18. In each of these samples, 32.5 wt% (based on the whole dispersion) of titanium dioxide (TiO_2), zinc oxide (ZnO), and alumina, taken individually, was dispersed in a hydrophobic solvent in the presence of 5 wt% of a polymeric surface modifier including a claimed copolymer and copolymers of the '945 patent. As may be noted from Table II below, all copolymers acted similarly, exhibiting what is typically expected for the effect of a steric stabilizer on dispersion rheology, inasmuch as they all reduced the viscosity of the TiO_2 dispersions, compared to a TiO_2 dispersion lacking a surface modifier, e.g., compare Sample No. 1 to Sample Nos. 4 and 7-10. As with a copolymeric surface modifier of the present invention, e.g., PEG 30 dipolyhydroxystearate, this expected behavior for a polymeric dispersant was observed even in ZnO and alumina dispersions (compare Sample Nos. 2 and 3 to Sample Nos. 5 and 6, respectively), albeit not in smectite clay dispersions (as noted in Table I of paragraph 16).

18.

Table II

Sample No.	Surface modifier	Solid, % by weight	Solvent	Brookfield Viscosity
1	None	TiO_2 , 32.5	Octyl stearate	51,855 cps @ 5 rpm
2	None	ZnO, 32.5	Octyl stearate	2,140 cps @ 5 rpm
3	None	Alumina, 32.5	Octyl stearate	60,000 cps @ 5 rpm
4	PEG 30 dipolyhydroxystearate (a copolymer of the present invention)	TiO_2 , 32.5	Octyl stearate	156 cps @ 5 rpm
5	PEG 30 dipolyhydroxystearate (a copolymer of the present invention)	ZnO, 32.5	Octyl stearate	68 cps @ 5 rpm
6	PEG 30 dipolyhydroxystearate (a copolymer of the present invention)	Alumina, 32.5	Octyl stearate	388 cps @ 5 rpm

7	Poly(butyl methacrylate-co-methyl methacrylate) (a copolymer from the first set of suitable copolymers cited in the '945 patent)	TiO ₂ , 32.5	Octyl stearate	4,410 cps @ 5 rpm
8	Poly(ethylene-co-methacrylic acid) (a copolymer from the second set of suitable copolymer cited in the '945 patent)	TiO ₂ , 32.5	Octyl stearate	18,920 cps @ 5 rpm
9	Poly(isoprene-graft-maleic anhydride) (a copolymer from the list of suitable graft copolymers cited in the '945 patent)	TiO ₂ , 32.5	Octyl stearate	144 cps @ 5 rpm
10	Polyglyceryl-2 dipolyhydroxystearate	TiO ₂ , 32.5	Octyl stearate	96 cps @ 5 rpm

19. The unexpected nature of the claimed compositions of the present invention is evident from yet another set of data presented in Table I and Table II. PEG 30-dipolyhydroxystearate is an amphipathic copolymer of the present invention. However, polyglyceryl-2 dipolyhydroxystearate, despite its partial resemblance with the said copolymer of the present invention, did not produce thickening in smectite clay dispersion (Sample No. 4, Table I). In contrast, this copolymer showed the expected steric stabilizing functionality in TiO₂ dispersion (Sample No. 10, Table II) by reducing the dispersion viscosity. To reflect further on the unexpected finding from the present invention, it might be relevant noting the effect of a water-soluble polymeric dispersant such as sodium polyacrylate on the rheology of aqueous dispersions of smectite clay and TiO₂, as presented in Table III of paragraph 20. (Note that smectite clays can thicken water or aqueous compositions without requiring any surface-modification.) This polymeric surface modifier, unlike the claimed copolymers of the present

invention, reduced the viscosity of not only the TiO₂ dispersion, but also of the smectite clay dispersion, as expected from a polymeric dispersant.

20.

Table III

Sample No.	Dispersed Solids, Wt%	Sodium Polyacrylate, Wt%	Brookfield Viscosity, 5 rpm
1	Smectite clay, 8 (dry basis)	0	49,600
2	Smectite clay, 8 (dry basis)	0.40	4,960
3	Titanium Dioxide, 32.4	0	24,000
4	Titanium Dioxide, 32.4	5	5,800

21. So in summary, with respect to the '945 patent, several commercially available copolymers of the types listed in the '945 patent were tested for their ability to modify the smectite clay surface in a manner where clay dispersions in hydrophobic liquids would show appreciable thickening. No thickening was observed. Yet, each of these copolymers exhibited the steric stabilizing functionality in TiO₂ dispersion by lowering the dispersion viscosity. A claimed amphoteric copolymer of the present invention also showed the expected behavior for a steric stabilizer inasmuch as it reduced the viscosities of titanium dioxide (TiO₂), alumina, and zinc oxide (ZnO) dispersions. In contrast, however, upon surface-treating smectite clay with the claimed copolymers, clay dispersions in hydrophobic liquids thickened significantly, albeit unexpectedly, considering that the same copolymer resulted in viscosity reductions in TiO₂, alumina, and ZnO dispersions. Based on the knowledge that a claimed copolymer serves as a steric stabilizer for TiO₂, alumina, and ZnO dispersions, a person skilled in the art would expect, at most, the copolymer to *reduce the viscosity of smectite clay dispersions*.

22. As noted in paragraphs 9 and 10, an unexpected benefit of the present invention is providing smectite clay-based thickeners for hydrophobic liquids by modifying the clay surface with a polymeric surface modifier, as opposed to a traditional QAC. To put the thickening ability of the claimed, copolymer-modified organoclays into a perspective, compositions thickened by a claimed organoclay were compared with compositions thickened by a QAC-modified organoclay for their viscosities. To conduct this comparative evaluation, eight additional samples were prepared under my supervision, which are summarized in Table IV of paragraph 23. These samples were prepared by diluting concentrated, gel-like dispersions of a purified smectite clay (having a cation exchange capacity of greater than 75 milliequivalents per 100 gram of clay, as required by the '218 patent) in octyl stearate (a hydrophobic liquid), with a given hydrophobic liquid. These gel-like dispersions had comparable levels (10.5 wt%) of the smectite clay, but the clay surface modifier was PEG 30 dipolyhydroxystearate (a copolymer of the present invention) for the concentrated dispersion used in preparing Sample Nos. 1-4, and methyl benzyl dihydrogenated tallow ammonium chloride (a QAC of the '218 patent) for the concentrated dispersion used in preparing Sample Nos. 5-8. The amount of the smectite clay in each of these dilute samples (i.e., Sample Nos. 1-8) was about 5 wt% (moisture-free basis). The dosage of the copolymer in Sample Nos. 1-4 was about 100 wt%, based on the weight of dry clay. The dosage of the QAC in Sample Nos. 5-8 was about 74 wt%, based on the weight of dry clay, which corresponds to about 115

milliequivalent per 100 grams of dry clay, falling midway in the optimum QAC dosage-range specified in the '218 patent, i.e., 100-130 milliequivalent per 100 grams of dry clay. All samples in Table IV below contained a polar activator, propylene carbonate, added at a dosage of about 35 wt%, based on the weight of dry clay.

23.

Sample No.	Dilutent	Concentrated Gel Dosage, g	Diluent Dosage, g	Brookfield Viscosity	
				1 rpm	5 rpm
Concentrated Gel in Octyl Stearate: Clay Surface modifier: PEG 30 Dipolyhydroxystearate					
1	Mineral Oil	71.43	78.57	364,000	196,000
2	Isododecane	71.43	78.57	120,000	56,000
3	Castor Oil	71.43	78.57	1,204,000	396,000
4	Dimethicone , 1.5 cst fluid	71.43	78.57	240,000	96,000
Concentrated Gel in Octyl Stearate: Clay Surface modifier: Methyl Benzyl Dihydrogenated Tallow Ammonium Chloride					
5	Mineral Oil	71.43	78.57	140,000	56,000
6	Isododecane	71.43	78.57	70,000	22,000
7	Castor Oil	71.43	78.57	400,000	112,000
8	Dimethicone , 1.5 cst fluid	71.43	78.57	140,000	40,000

24. As an additional comment concerning the '218 patent, cationic surfactants, e.g., QACs, adsorb only onto the face-surface of smectite clay platelets via an ion-exchange process. Accordingly, the adsorption of these surfactants onto the clay surface relies on the cation exchange capacity (CEC) of the clay. For this reason, the '218 patent specifies a minimum CEC value (75 milliequivalents per 100 gm of clay) for the '218 invention to work. This restriction

minimum CEC value (75 milliequivalents per 100 gm of clay) for the '218 invention to work. This restriction does not apply in the case of the presently claimed composition, because the adsorption of the copolymer(s) of the present invention onto the clay surface is not via an ion-exchange process. Clearly, the '218 patent teachings are limited to an ion exchange-based clay treatment requiring, indispensably, a cationic surface modifier, selected from QACs, and absolutely fails to teach or suggest any other monomeric, let alone polymeric, surface-modifying agent for the smectite clay. The QAC disclosed in the '218 patent is a monomeric, low molecular weight cationic surfactant, as opposed to the polymeric surface-modifying compounds recited in the present claims, which are not cationic. The '218 patent, therefore, cannot possibly teach or suggest the specific type of copolymer recited in the present claims.

25. All statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true; further, these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001, Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or document or any patent resulting therefrom.

Ashoke K. SenGupta
Ashoke K. SenGupta

Dated: January 16, 2006